

Unconventional charge density wave in the organic conductor α -(BEDT-TTF)₂KHg(SCN)₄

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The low temperature phase (LTP) of α -(BEDT-TTF)₂KHg(SCN)₄ salt is known for its surprising angular dependent magnetoresistance (ADMR), which has been studied intensively in the last decade. However, the nature of the LTP has not been understood until now. Here we analyse theoretically ADMR in unconventional (or nodal) charge density wave (UCDW). In magnetic field the quasiparticle spectrum in UCDW is quantized, which gives rise to spectacular ADMR. The present model accounts for many striking features of ADMR data in α -(BEDT-TTF)₂KHg(SCN)₄.

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The series of quasi-two dimensional organic conductors α -(BEDT-TTF)₂MHg(SCN)₄ (where BEDT-TTF denotes bis(ethylenedithio)tetrathiafulvalene and M=K, NH₄, Rb and Tl) have attracted considerable attention over the last few years due to two different ground states and rich phenomena associated with them[1].

Whereas the M=NH₄ compound becomes superconducting below 1.5K, other salts enter at $T_c = 8 - 12$ K into a specific low temperature phase (LTP) with striking ADMR. From the magnetic phase diagram of LTP it is now believed that LTP is not SDW but a kind of CDW, though no detailed characterization is available[2]. We have proposed recently that unconventional (or nodal) charge density wave (UCDW) can account for a number of features in LTP in α -(BEDT-TTF)₂KHg(SCN)₄ including the threshold electric field[3, 4, 5, 6, 7]. Recently UCDW and USDW have been proposed by several authors as possible electronic ground state in quasi-one dimensional and quasi-two dimensional crystals[8, 9, 10, 11, 12]. Unlike conventional DW[13], the order parameter in UCDW $\Delta(\mathbf{k})$ depends on the quasiparticle wave vector \mathbf{k} . In α -(BEDT-TTF)₂KHg(SCN)₄ salts, where the conducting plane lies in the a-c plane and the quasi-one dimensional Fermi surface is perpendicular to the a-axis, we assume that $\Delta(\mathbf{k}) = \Delta \cos(ck_z)$ or $\Delta \sin(ck_z)$ (i.e. $\Delta(\mathbf{k})$ depends on \mathbf{k} perpendicular to the most conducting direction), where $c = 9.778\text{\AA}$ is the lattice constant along the c-axis[14]. It is known also that the thermodynamics of UCDW and USDW is practically the same as the one in d-wave superconductor[11, 15]. Also in spite of the clear thermodynamic signal, the first order terms in $\Delta(\mathbf{k})$ usually vanishes when averaged over the Fermi surface. This implies neither clear x-ray signal for UCDW, nor spin signal for USDW. Due to this fact unconventional density waves are sometimes called the phase with hidden order parameter[12].

In a magnetic field the quasiparticle spectrum is quantized as first shown by Nersesyan et al.[8, 9]. This dramatic change in the quasiparticle spectrum is most readily seen in ADMR as it has been demonstrated recently for SDW plus USDW in (TMTSF)₂PF₆ below $T = T^*(\sim 4\text{K})$ [16]. About a decade ago ADMR in LTP in α -(BEDT-TTF)₂KHg(SCN)₄ salts have been studied intensively. In particular ADMR for current \mathbf{j} perpendicular ($\mathbf{j} \parallel \mathbf{b}^*$) and parallel to the a-c plane exhibits a broad peak around $\theta = 0^\circ$ (see insert in Fig. 1), where θ is the angle with which the magnetic field is tilted from the b-axis (normal to the conducting plane). In addition, a series of dips are observed

at $\theta = \theta_n$ given by[17, 18]

$$\tan(\theta_n) \cos(\phi - \phi_0) = \tan(\theta_0) + nd_0, \quad (1)$$

where $\tan \theta_0 \simeq 0.5$, $d_0 \simeq 1.25$, $\phi_0 \simeq 27^\circ$ and $n = 0, \pm 1, \pm 2, \dots$. Here ϕ is the angle the projected magnetic field on the a-c plane makes with the c-axis. The origin of this surprising ADMR have been discussed but apparently without clear answer[17, 18, 19, 20, 21, 22]. In the following we shall show that the quasiparticle spectrum in UCDW in α -(BEDT-TTF)₂KHg(SCN)₄ salts is quantized in the presence of magnetic field. The small energy gap which is proportional to \sqrt{B} where B is the field strength, depends also on the direction of the magnetic field and it can be seen in ADMR. As it will be shown below, we can describe salient aspects of ADMR seen in LTP of α -(BEDT-TTF)₂KHg(SCN)₄ very consistently. Therefore we may conclude that ADMR in α -(BEDT-TTF)₂KHg(SCN)₄ provides definitive evidence that LTP is UCDW. We stress that the Landau quantization as proposed by Nersesyan et al.[8, 9] should be readily accessible in other UCDW and USDW systems. In this respect experimental analysis of ADMR in the pseudogap phase in high T_c cuprate superconductors[23] and the glassy phase in κ -(BEDT-TTF)₂Cu[N(CN)₂]Br salt[24] will be of great interest. In α -(BEDT-TTF)₂KHg(SCN)₄ salts the conducting plane is the a-c plane and the quasi-one dimensional Fermi surface is perpendicular to the a-axis. In addition there is a quasi-two dimensional Fermi surface with elliptical cross section in the a-c plane. In LTP we assume that UCDW appears on the quasi-one dimensional Fermi surface with quasiparticle energy given by

$$E(\mathbf{k}) = \sqrt{\xi^2 + \Delta^2(\mathbf{k})} - \varepsilon_0 \cos(2\mathbf{b}'\mathbf{k}), \quad (2)$$

where $\xi \approx v_a(k_a - k_F)$, $\Delta(\mathbf{k}) = \Delta \cos(ck_c)$ and ε_0 is the parameter describing the imperfect nesting[25, 26, 27, 28]. In fitting the experimental data we discovered that 1. Eq. (2) gives only one single dip in ADMR, 2. therefore the imperfect nesting term has to be generalized as

$$\varepsilon_0 \cos(2\mathbf{b}'\mathbf{k}) \longrightarrow \sum_n \varepsilon_n \cos(2\mathbf{b}'_n\mathbf{k}), \quad (3)$$

where $\mathbf{b}'_n = b'(\cos \theta_n \hat{\mathbf{k}}_b + \sin \theta_n (\hat{\mathbf{k}}_a \cos \phi_0 + \hat{\mathbf{k}}_c \sin \phi_0))$ and $\varepsilon_n \sim 2^{-|n|}$. Eq. (3) indicates that the imperfect nesting term does not follow from a usual tight binding model but appears to have an interesting superstructure whose meaning is not clear at this moment. As seen from Eq. (2), the quasiparticle spectrum is gapless and LTP is metallic in sharp contrast to conventional CDW. In a magnetic field the first term of the quasiparticle spectrum changes to

$$E_n = \pm \sqrt{2nv_a \Delta c e |B \cos \theta|}, \quad (4)$$

where $n = 0, 1, 2, \dots$. This is readily obtained following Refs. 8, 9. The contribution from the imperfect nesting term is considered as a perturbation and the lowest order corrections to the energy spectrum are given by:

$$E_0^1 = E_1^1 = - \sum_m \varepsilon_m \exp(-y_m), \quad (5)$$

$$E_1^2 = - \sum_m \varepsilon_m (1 - 2y_m) \exp(-y_m), \quad (6)$$

where $y_m = v_a b'^2 e |B \cos(\theta)| (\tan(\theta) \cos(\phi - \phi_0) - (\tan(\theta_0) + md_0))^2 / \Delta c$. The $n = 1$ level was twofold degenerate, but the imperfect nesting term splits the degeneracy by E_1^1 and E_1^2 . Also the imperfect nesting term breaks the particle-hole symmetry. When $\beta E_1 \gg 1$ ($\beta = (k_B T)^{-1}$), the quasiparticle transport in the quasi-one dimensional Fermi surface is dominated by the quasiparticles at $n = 0$ and $n = 1$ Landau levels. Considering that there are 2 conducting channels and only the quasi-one dimensional one is affected by the appearance of UCDW, the ADMR is written as

$$R(B, \theta, \phi)^{-1} = 2\sigma_1 \left(\frac{\exp(-\beta E_1) + \cosh(\beta E_1^1)}{\cosh(\beta E_1) + \cosh(\beta E_1^1)} + \frac{\exp(-\beta E_1) + \cosh(\beta E_1^2)}{\cosh(\beta E_1) + \cosh(\beta E_1^2)} \right) + \sigma_2 \quad (7)$$

Here σ_1 and σ_2 are the conductivities of the $n = 1$ Landau level and quasi-two dimensional channels, in which the contribution of the $n = 0$ Landau level was melted, respectively. In Figs. 1 and 2 we compare the B dependence of the magnetoresistance at $T = 1.4\text{K}$ and $T = 4.14\text{K}$ and the T dependence of the magnetoresistance for $B = 15\text{T}$ for $\theta = 0$. In fitting the temperature dependence of the resistivity, we assumed $\Delta(T)/\Delta(0) = \sqrt{1 - (T/T_c)^3}$, which was found to be very close to the exact solution of $\Delta(T)$ [11].

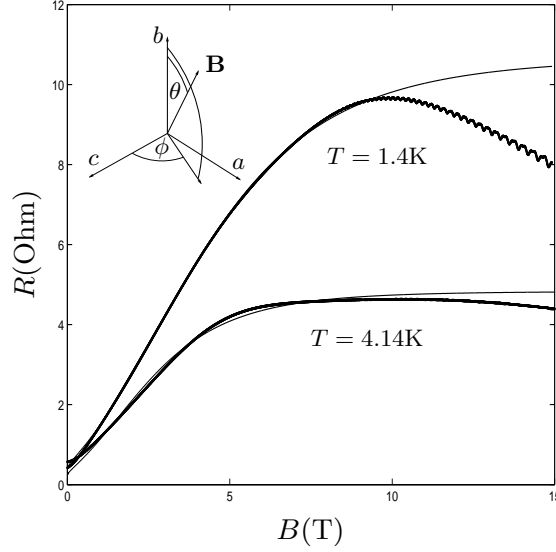


FIG. 1: The magnetoresistance is plotted for $T = 1.4\text{K}$ and 4.14K as a function of magnetic field. The thick solid is the experimental data, the thin one denotes our fit based on Eq. (7). The inset shows the geometrical configuration of the magnetic field with respect to the conducting plane.

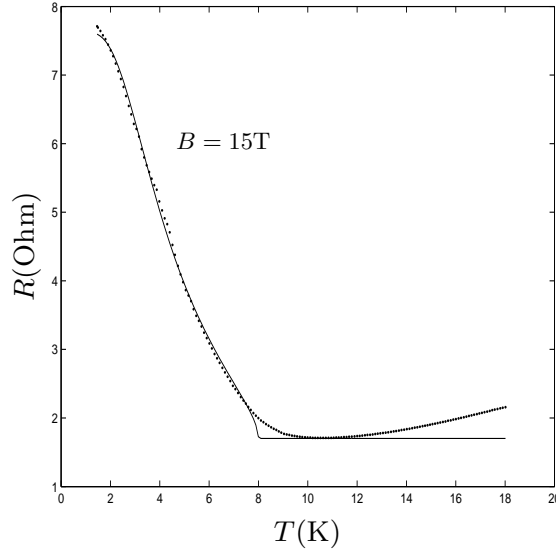


FIG. 2: The temperature dependent magnetoresistance is shown at $B = 15\text{T}$. The dots are the experimental data, the solid line is our fit.

Clearly the fitting becomes better as T decreases and/or B increases. Also for $T = 1.4\text{K}$ Shubnikov-de Haas oscillation becomes visible around $B = 10\text{T}$, then the fitting starts breaking away. Clearly in this high field region the quantization of Fermi surface itself starts interfering with the quantization described above. In this region, the explicit B and T dependence of σ_1 and σ_2 should be taken into account what we neglected here for simplicity. Also the deviation of the theoretical curve from the experimental one above T_c in Fig. 2 is originated from this neglect. Here we concentrated on the dominant conduction mechanism, that is thermally excited quasiparticles across the magnetic field induced gap. From these fittings we obtain σ_2/σ_1 of the order of 0.1, and by assuming the mean field value of Δ (17K), we get v_a of the order of 10^6cm/s . In Figs. 3 and 4 we show the experimental data of ADMR as a function of θ for current parallel and perpendicular to the conducting plane for $T = 1.4\text{K}$, $B = 15\text{T}$ and $\phi = 45^\circ$. As is readily seen the fittings are excellent. From this we deduce σ_2/σ_1 of the order of 0.1, $\varepsilon_0 = 4.2\text{K}$, b' is of the order of a few lattice constants from these fittings. Finally we show in Fig. 5 R versus θ for different ϕ and compare with the experimental data side by side. Perhaps there are still differences in some details but the overall agreement is very

striking. These differences might arise from the fact, that similarly to the neglect of magnetic field and temperature dependence of σ_1 and σ_2 , we also assumed them to be independent of ϕ and θ . The present model can describe a similar figure found in Ref. 20 rather well.

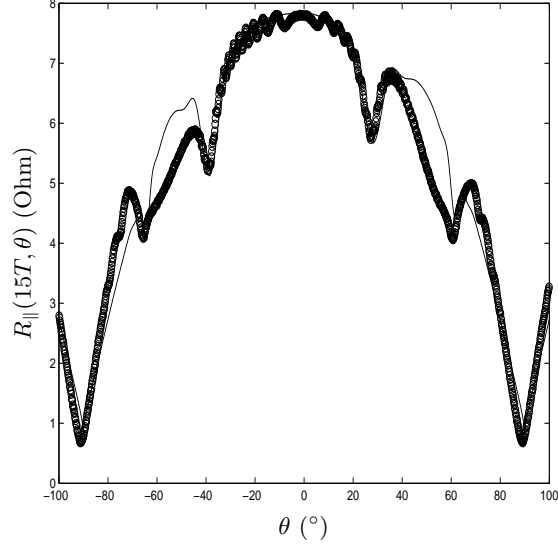


FIG. 3: The angular dependent magnetoresistance is shown for current parallel to the a-c plane at $T = 1.4\text{K}$, $B = 15\text{T}$. The open circles belong to the experimental data, the solid line is our fit based on Eq. (7).

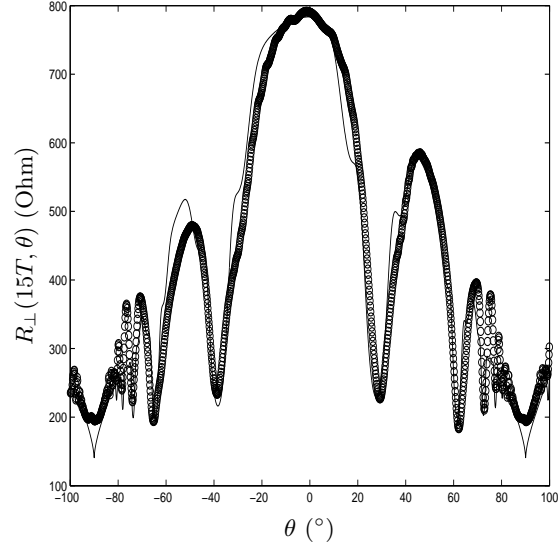


FIG. 4: The angular dependent magnetoresistance is shown for current perpendicular to the a-c plane at $T = 1.4\text{K}$, $B = 15\text{T}$. The open circles belong to the experimental data, the solid line is our fit from Eq. (7).

In summary we have succeeded in describing the salient feature of ADMR observed in LTP in $\alpha\text{-(BEDT-TTF)}_2\text{KHg(SCN)}_4$ in term of UCDW with the Landau quantization of the quasiparticle spectrum. Very similar ADMR have been seen in M=Rb and Tl compounds as well. Therefore we conclude that LTP in $\alpha\text{-(BEDT-TTF)}_2\text{MHg(SCN)}_4$ salts should be UCDW. Also we believe that ADMR provides clear signature for the presence of UCDW and USDW. Therefore this technique can be exploited for other possible candidates of UDW.

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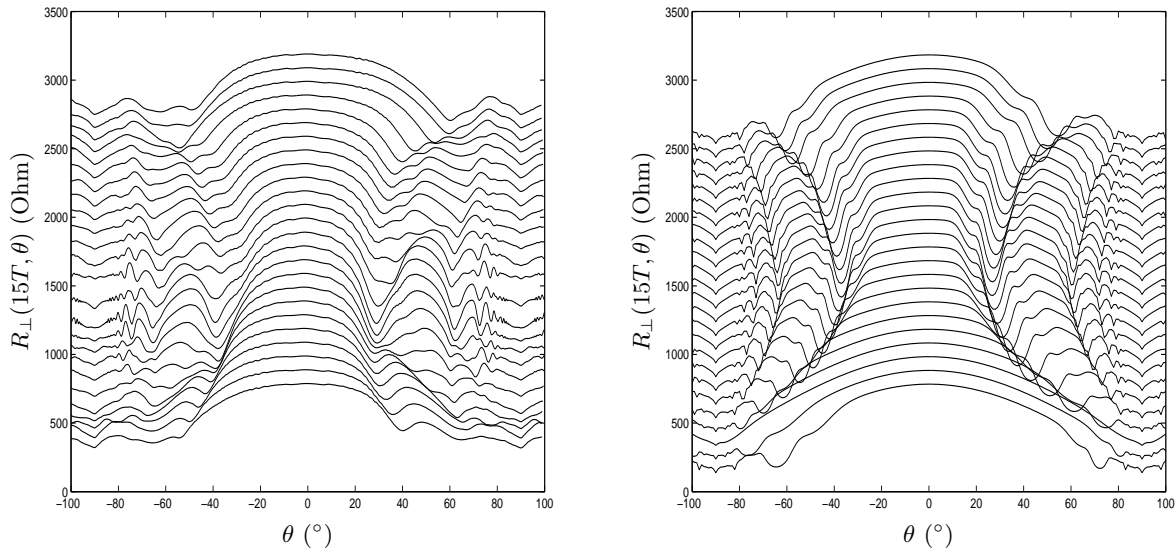


FIG. 5: ADMR is shown for current perpendicular to the a-c plane at $T = 1.4\text{K}$ and $B = 15T$ for $\phi = -77^\circ, -70^\circ, -62.5^\circ, -55^\circ, -47^\circ, -39^\circ, -30.5^\circ, -22^\circ, -14^\circ, -6^\circ, 2^\circ, 10^\circ, 23^\circ, 33^\circ, 41^\circ, 48.5^\circ, 56^\circ, 61^\circ, 64^\circ, 67^\circ, 73^\circ, 80^\circ, 88.5^\circ, 92^\circ$ and 96° from bottom to top. The left (right) panel shows experimental (theoretical) curves, which are shifted from their original position along the vertical axis by $n \times 100\text{Ohm}$, $n = 0$ for $\phi = -77^\circ$, $n = 1$ for $\phi = -70^\circ$,

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